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Citation: Journal of Applied Physics 40, 2817 (1969); doi: 10.1063/1.1658081
View online: http://dx.doi.org/10.1063/1.1658081
View Table of Contents: http://scitation.aip.org/content/aip/journal/jap/40/7?ver=pdfcov
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Magnetic Resonance Studies of Lithium Vanadium Bronze
Magnetization Studies on Superconducting Vanadium-Gallium

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(Received 22 November 1968; in final form 20 February 1969)

The temperature dependence of the critical fields of V₃Ga has been measured up to 70 kOe using ac and dc magnetization techniques. For a material with a transition width of 26 mdeg at a transition temperature, $T_\text{c}$ of 14.19°K, one calculates from the slope $(dH_c/dT = -43.2 \, \text{kOe/deg})$ and curvature of the $H_c$ vs $T$ curve, the Ginzburg-Landau parameter, $\kappa = 40 \pm 2$, the Pauli paramagnetism parameter $\alpha = 2.28 \pm 0.04$, and the spin-orbit scattering constant $\lambda_{so} = 0.6 \pm 0.1$. No evidence for $H_a$ was detected and the experimentally difficult upper limit to $H_a$ appears to be about three times as large as calculated from the theories.

I. INTRODUCTION

The intermetallic compound V₃Ga and its superconductivity were discovered by Matthias, Wood, Corenzwit, and Bala.¹ The material is a high-field, type II superconductor with transition temperatures $T_\text{c}$ ranging from 14.2° to 16.8°K. These variations may be caused by slight differences in composition and in heat treatment during or after formation of the compound. Several properties of superconducting V₃Ga have been measured near $T_\text{c}$ and observations of the upper critical field over an extended temperature range have also been published.² This work indicates, as also noted by the present authors,³ that the experimental upper critical field, $H_{c2}$, is reduced below the Ginzburg-Landau-Abrikosov-Gorkov (GLAG) upper critical field, $H_{c2}^*$, by the effect of Pauli spin paramagnetism in the normal state.⁴ However, recent experimental⁵ and theoretical⁶ studies suggest that for some high-field superconductors spin-orbit scattering in the superconducting state may partially offset the effect of Pauli spin paramagnetism.

Both dc and high sensitivity ac techniques were used to determine the magnetic properties of hollow cylinders of superconducting V₃Ga in dc magnetic fields up to 70 kOe. The magnetization curves are similar to those of other tubular samples of type II superconductors as measured by Kim, Hempstead, and Strnad³ and interpreted by Anderson’s⁴ flux creep mechanism. Since, according to this model the motion of flux cannot be instantaneous, slight ac frequency effects should be and have been observed. However, the present paper is limited to “steady-state” magnetization curves, critical currents, the various critical fields, and to an analysis of their temperature dependence, leaving frequency effects on flux motion to another article.

II. EXPERIMENTAL

The vanadium-gallium samples studied in this work were obtained as small arc-melted buttons from Professor B. T. Matthias. They were used as received with no further heat treatment, but there were variations in transition temperatures and widths. We selected the sample with the narrowest transition as being indicative of the greatest homogeneity. The sample was fabricated by spark cutting into a hollow cylinder 8.13-mm long, 2.49-mm o.d., and 1.70-mm i.d.

The experiments were performed with the Los Alamos liquid-hydrogen-cooled solenoid⁷ capable of producing 80 kOe in a 5-cm-diam low-temperature core.

The field calibration was accurate to better than 1%. The magnetic field was controlled by an integrating feedback circuit⁸ and could be held constant or swept at any desired rate up to 10 kOe/sec.

Two cryostats were used. The first was a simple liquid-hydrogen bath in which the temperature was taken from the hydrogen vapor pressure, using the equation given by Weber et al.,⁹ based upon the NBS-1955 scale. The second apparatus was a thermally floating helium-cooled copper capsule capable of operating in the temperature range between 2° and 30°K. The temperature of the capsule could be maintained

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¹⁵ H. L. Laquer, in Ref. 3, p. 156.
stable to within ±0.002°K using an electronic bridge controller. The temperature of the sample was measured with an Allen–Bradley 0.1 W, 43-Ω carbon resistor. This resistor was calibrated using points in the helium vapor pressure range and it was also compared in situ with a germanium thermometer subsequently calibrated at the U. S. National Bureau of Standards. The magnetoresistance of the carbon thermometer was measured in liquid helium and in liquid hydrogen and appropriate corrections were applied at intermediate temperatures. At 68 kOe and 12.2°K these corrections were equivalent to 79 mdeg. Our overall temperature accuracy is estimated to be ±20 mdeg and the precision which enters in determining slopes ±10 mdeg.

The V$_2$Ga cylinder was located inside a primary or drive coil which could be made to generate an ac field of 0.01 to 60 Oe peak to peak (p-p) for frequencies in the range from 40 Hz to 400 kHz. Inside the cylinder was a pickup coil, about 3 mm long wound with 500 turns of B & S #49 gauge (0.0025-cm-diam) Formex insulated copper wire. This assembly could be placed in either of the cryostats and positioned in the center of the high-field solenoid. The ac frequency and magnitude were preset and the signal from the pickup coil (amplified on a broad-band Hewlett Packard model 3400A voltmeter) was recorded on an X–Y recorder at a given temperature while sweeping the dc field, or at a given dc field while slowly changing the temperature. Maximum noise level was always less than 10% of the normal state signal.

For the dc measurements two matched coils were used, one of which closely surrounded the V$_2$Ga sample. The coils were connected in opposition and the difference of the induced voltages was integrated by a Dymec model 2460A operational amplifier to give the amount of flux excluded from the coil containing the specimen. This difference was plotted against the magnetic field on an X–Y recorder. Due to residual thermal emf’s in the circuit and the small size of our sample it was only possible to detect field differences greater than 1 G.

III. RESULTS

A typical zero-field transition using a 0.05 Oe (p–p), 100-kHz signal and sweeping the temperature at 0.002°K/min is shown in Fig. 1. This transition is completely reversible with less than 0.002°K hysteresis. The full width of the transition from the point where the normal signal has dropped 0.5% to the point where the drop is 99.5% complete amounts to 0.056°K. This value represents an upper limit. A more generally accepted width value is obtained as the distance between the intercepts with the horizontal lines of the steep linear region of the transition curve (between the 1/4 and 3/4 completion points). This method of interpretation of the experimental data then yields a transition width of only 26 mdeg.

Several measurements of the normalized ac output signal against the dc field at constant temperature are shown in Fig. 2. These curves are also completely reversible, as long as the field is swept slowly so as to avoid heating the sample. Straight-line extrapolation gives a width of about 2.4 kOe for the transition as measured at the 0.05-Oe p–p ac level and noticeably greater values at the higher ac levels.

Fig. 1. Zero field transition at 100 kHz, 0.05 Oe p–p. Ac signal E from pickup coil as a function of carbon resistor thermometer voltage, mV, at a fixed measuring current of 103.5 μA (lower scale) and corresponding temperature T (upper scale). Steps are caused by 1-sec sampling period of digital voltmeter used for carbon thermometer readout.

Fig. 2. Transitions in dc magnetic field for various ac levels at 13.52°K. Normalized ac signal from pickup coil, $E/E_N$, where $E_N$ is the normal state limiting value of the ac signal $E$ plotted as a function of the dc field $H$ for various indicated values of the alternating field intensity.

19 J. D. Lindsay, Rev. Sci. Instrum. 37, 1192 (1966).
Taking 0.5% changes in this output signal as reproducible decision levels, we obtain field values, $H_N$ for the normal and $H_s$ for the superconducting limit of the transition. Figure 3 shows the relation between $H_s$ and the magnitude of the applied ac field. These are essentially critical current curves. For any given temperature there is a break or knee, indicated by the arrows, defining a field $H_K$. $H_N$ also varies somewhat with the magnitude of the ac field, its maximum value $H_{N\text{ (max)}}$ being obtained with the smallest ac field. Since the midpoint field, where the output signal is one half of the normal state signal, can be determined with greater accuracy than either $H_K$ or $H_N$, the field at the midpoint of the 100 kHz and 0.05-Oe (p-p) transitions was used to analyze the temperature dependence of the upper critical field and is thus designated as $H_{c2}$. The temperature variations of these various fields are shown in Fig. 4.

The dc measurements gave curves as shown in Fig. 5. The point of deviation from the Meissner line for the virgin curve, as indicated by the arrow in this figure, represents a maximum value for the lower critical field $H_{c1}$ divided by $(1 - n)$, where $n = 0.2$ is our estimate of the demagnetizing factor. These experimental values of $H_{c1}$ are plotted in Fig. 6. The slope of the dc magnetization curve near its upper end was so small that an accurate value of $H_{c1}$ could not be determined. The width of the hysteresis, as in Fig. 5, is a measure of the maximum shielding currents and hence can be plotted with the ac critical current measurements in Fig. 3. Comparison between the ac and dc measurements is quite satisfactory in the range of validity of each.

**IV. DISCUSSION**

The measured zero-field transition width of 26 mdeg is narrower, by a factor of 72, than that predicted by Pippard’s thermodynamic fluctuation theory for this material with a coherence length of 25 Å (see below). Actually, the true fluctuation-limited transition width is probably smaller than our measurements by a factor of 300 since the coherence length approaches the sample dimension at $T_e$ as pointed out by Shier and Ginsberg. Nevertheless, considering the type of material, our measured width indicates an acceptable homogeneity for the V$_2$Ga cylinder used in most of this study.

The dc magnetization curves are similar to those of other type II materials. The hysteresis pattern is symmetrical about the zero magnetization line because the specimen is multiply connected. Hence the hysteresis does not by itself imply irreversibility of the material. However, the absence of a sharp drop at the peak near $H_{c1}$ as observed in reversible type II super-
coherence distance and \( l = 12 \, \text{Å} \) is the electron mean free path estimated from the normal state resistivity \( \rho_n = 36 \, \mu\Omega \cdot \text{cm} \). The surface layer under these conditions would carry a current density of 160 A/cm².

At any rate, since the slopes are the same, we have no evidence for surface superconductivity in our specimen which leaves us with the following possibilities: (a) there is no surface nucleation in V₃Ga, (b) the net current capacity of the surface layer is less than 160 A/cm², or (c) more specialized surface preparation methods than presently available are needed to exhibit surface superconductivity in V₃Ga.

Since the temperature dependence of all the curves in Fig. 4 is essentially the same, we are justified in choosing the most accurately measured one, \( H_{c2} \), to calculate other properties of V₃Ga. The upper critical field increases so rapidly near \( T_c \) that our measurements only extend to a reduced temperature of 0.86. This range and the precision of our data, however, are sufficient to determine both the initial-slope and curvature of the upper critical field versus temperature curve, and hence the parameters \( \alpha \) and \( \lambda_0 \) which have been used in recent theories to characterize type II superconductors.

The first of these,

\[
\alpha = \sqrt{H_{c2}^*(0)/H_T(0)},
\]

was defined by Maki in his theory of the upper critical field for a “dirty” superconductor including the effects of Pauli spin paramagnetism in the normal state, where \( H_T(0) \), introduced by Clogston, represents the hypothetical critical field in the absence of all magnetization effects other than spin paramagnetism, and \( H_{c2}^*(0) \) the GLAG upper critical field, both at the absolute zero.
The second or spin-orbit scattering parameter $\lambda_{so}$ was introduced by Werthamer et al.\(^{23}\) in their more general treatment which includes the compensatory paramagnetic lowering of the free energy of the superconducting state due to spin orbit scattering. They give

$$-\ln t = \sum_{n=0}^{\infty} \left| \frac{2n+1}{t} \right|^{-1} \left[ \frac{h^2}{2n+1} + \frac{h}{t} + \left( \frac{a^2h^2}{t} \right) \right], \quad (2)$$

with the dimensionless variables $t$, $h$, $a$, and $\lambda_{so}$ as defined in Eq. (27) of their paper.

We have obtained an expansion of Eq. (2) valid for $T$ near $T_e$ in which we let $t=1-T/T_e<1$. This expansion to order $t$ yields

$$h = A' t (1-B t), \quad (3)$$

with

$$A' = \pi^2/4 \quad (4)$$

and

$$B = \frac{3}{2} - \left( \frac{28}{\pi^4} \right) t + \left( 16a^2/\pi^4 \lambda_{so} \right)$$

$$\times \left[ \frac{1}{4} \pi^2 - \lambda_{so} \left( \psi \left( \frac{3}{2} \lambda_{so} \right) + \frac{1}{2} \right) - \psi \left( \frac{3}{2} \right) \right], \quad (5)$$

where the Riemann function $\xi(3)=1.202$ and $\psi(z)$ is the digamma function.\(^{26}\)

There are two methods of obtaining $\alpha$ discussed by Werthamer et al.\(^{23}\) One follows from the experimental slope of $H_{cr}$ near $T_e$

$$\alpha = 5.2758 \times 10^{-5} \left( -dH_{cr}/dT \right)_{T_e}, \quad (6)$$

and the other involves experimental data on the normal state, which in the short mean-free path or "dirty" limit gives

$$\alpha = 3 \pi^2 \rho_s \kappa_{so} / 2 m^* k_{B}^2, \quad (7)$$

where $\gamma$ and $\rho_s$ are the normal state electronic specific heat and electrical resistivity, respectively, and the remaining quantities are fundamental constants. The value of $\gamma = 3.04 \times 10^9$ erg cm$^{-2}$ deg$^{-1}$ is taken from Morin et al.\(^{21}\) and $\rho_s = 36 \mu \Omega$ cm from Sarachik et al.\(^{28}\)

Our experimental data can be fitted to an equation of the form

$$H_{cr}(T) = A \theta (1-B \theta) \quad (8)$$

by a least-squares analysis with $A = 612 \pm 8$ kOe, $B = 1.27 \pm 0.10$, and $T_c = 14.16 \pm 0.01^\circ$K. This value of $T_e$ is 0.03$^\circ$K below the measured value of 14.19$^\circ$K because of a narrow region of positive curvature for $T_e/T_c > 0.985$ which cannot be accommodated by Eq. (8). The parameters reproduce the measured field values with an rms deviation of 0.55 kOe. From these experimental values of $A$, $B$, and $T_e$ the slope of $H_{cr}$ at $T_e$, $\alpha$, and $\lambda_{so}$ are obtained using Eqs. (5) and (6).

### Table I. Summary of results.

| $T_c$(meas) | 14.19$\pm 0.02$ K |
| $H_p(0)$ | 260 kOe |
| $H_{cr}(T_e)$ | $-43.2 \pm 0.7$ kOe/$^\circ$K |
| $H_{up}(0)$ | 420 kOe |
| $H_{cr}(0)$ | $-39.1 \pm 0.7$ kOe |
| $H_{up}(0)$ | 53 kOe |
| $\rho_s$ | $36 \mu \Omega$ cm |
| $\lambda_{so}$ | $0.6 \pm 0.1$ |
| $\kappa_{so}$ | $40 \pm 2$ |
| $\lambda_{so}$ | $48 \pm 5$ |

The results are given in Table I with the statistical errors representing one mean deviation.

Our initial slope near $T_c$ of $-43.2 \pm 0.7$ kOe/deg can be compared with Wernick et al.'s\(^{25}\) $-53$ kOe/deg and Montgomery and Wiggall's\(^{29}\) $-39.1$ kOe/deg both from direct resistance measurements and Morin et al.'s\(^{25}\) $-48$ kOe/deg from specific heat data. The value of $\kappa$ can also be estimated in two ways, both, however, involving the normal state electronic specific heat. From the definition $H_{cr}(0)=\lambda_{so} H_e^2$, and the slope $dH_{cr}/dT_e = 4.405 \gamma^{1/2}$ given by BCS\(^{27}\) one finds

$$\kappa = 0.1605 \gamma^{1/2} (-dH_{cr}/dT_e)_{T_e} \quad (9)$$

Goodman\(^{32}\) and Berlincourt\(^{33}\) estimate a value of $\kappa$ from the electronic properties of the normal metal. The major contribution arises from

$$\kappa_1 = 7.53 \times 10^9 \rho_s \gamma^{1/2} \quad (10)$$

where $\rho_s$ is in $\Omega$ cm. The results for $\kappa$ are also tabulated in the table.

A limiting expression for Eq. (2) valid at $T=0$ is given by Werthamer et al.\(^{23}\) as

$$2.54 = -\ln K + \left( \lambda_{so} / \gamma \right) \tan^{-1} \left[ \gamma / (h_0 + \lambda_{so}) \right], \quad (11)$$

where $K = (1 + a^2) h_0^2 + \lambda_{so} h_0$, $\gamma = (\gamma^2 - 1/2) \gamma_{so}^{1/2}$, and $h_0 = H_e(0) / 3.56 H_e^*(0)$.\(^{34}\) If we take $H_p(0) = 18400 T_e$,\(^{20}\) and $\alpha$ calculated from experimental results near $T_c$, the value of $H_{cr}(0)$ can be calculated from Eq. (1). Then from the values of $\lambda_{so}$ and $\alpha$ determined near $T_c$ we use Eq. (11) to calculate $H_{cr}(0)$. These results are given in the table. Our calculated $H_{cr}(0)$ of $270 \pm 60$ kOe is somewhat larger than the 208$\pm 5$ kOe reported by Montgomery and Wiggall\(^{29}\) from measurements at $4^\circ$K on very similar material. However, the agreement is within the estimated errors, which is quite satisfactory, considering the limitations of the present theories, the small range of reduced temperatures covered in our measurements and the extent of our extrapolation.

Finally, using the experimental value for $\kappa$ and Maki's\(^{29}\) expression for the lower critical field, $H_{cr}(T)$ one can calculate $H_{cr}(T)$ and compare it with the results from the dc magnetization measurements. This comparison is shown by the calculated curve for $H_{cr}$ in Fig. 6. It should be noted that the calculated $H_{cr}$ is less than

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\(^{23}\) B. B. Goodman, IBM J. Res. Develop. 6, 63 (1962).
1/3 of the experimental points. This discrepancy may well be due to the difficulty of determining the break from the linear Meissner region due to trapped shielding currents which cause the experimenter to overestimate $H_{el}$. Even the results on 70-μ powders by Swartz et al. gave an $H_{el}$ lower than our experimentally estimated $H_{el}$, but greater than the calculated value.

V. CONCLUSIONS

The ac method of measuring magnetization and the interpretation thereof described in the paper is in good agreement with more standard dc techniques but is considerably more sensitive in the region near $H_{el}$ where the magnetic moment is small. Given material with a sufficiently narrow transition, and thermometry of sufficient accuracy, our technique yields values for the slope as well as for the initial curvature of the upper critical field as a function of the temperature. These experimental numbers are then used together with the current theories to calculate more derived quantities, including a reliable value for the parameter $\alpha$, and an estimate for the spin orbit scattering parameter in the superconducting state.

The dc measurements emphasize the great difficulty of accurately determining $H_{el}$ in a massive specimen of nonideal type II superconductor in which there are considerable shielding currents in the region above $H_{el}$.

It is conceivable that there may be metallurgical differences between $V_{7}Ga$ samples used by different investigators. Nevertheless, wherever a comparison is possible, the properties of the $V_{7}Ga$ used in this study are not significantly different from those reported by others.

ACKNOWLEDGMENTS

We wish to thank Professor B. T. Matthias for supplying the $V_{7}Ga$ samples and encouraging us in this work and Dr. L. Gruenberg for a number of enlightening discussions. One of us also is grateful to Dr. E. F. Hammel and the cryogenics group at Los Alamos for the hospitality extended to him there as a Visiting Staff Member during his sabbatical leave.